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Modality in ambient particle size distributions and its potential as a basis for developing air quality regulation

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ABSTRACT

Current ambient air quality standards are mass-based and restricted to $PM_{2.5}$ and PM_{10} fractions. The major contribution to both $PM_{2.5}$ and PM_{10} fractions is from particles belonging to the coarse mode and generated by mechanical processes. These standards are thus unable to effectively control particle concentrations from combustion sources, such as motor vehicles and power plants, which tend to emit very small particles that

are almost entirely respirable and in the submicron range, and dominate the nucleation and accumulation modes, which contribute much less to particle mass concentration.

The aim of this work was to examine whether PM_1 and PM_{10} would be a more effective combination of mass standards than $PM_{2.5}$ (dominant in the nucleation and accumulation modes) and PM_{10} (dominant in the coarse mode) in controlling combustion related ambient particles, as well as those originating from mechanical processes. Firstly, a large body of data on particle size distributions in a range of environments in South East Queensland, Australia was analysed, with an aim of identifying the relation between modality in the distributions and sources of particles belonging to different modes. The analyses included a matrix of the following elements: particle volume and number distributions, type of environment and locations of the modes in the range of PM_1, $PM_{2.5}$ and PM_{10} fractions. Secondly, with the same aim, 600 published modal location values relating to number, surface area, volume and mass size distributions for a range of environments worldwide, were analysed. The analysis identified a clear and distinct separation between the location of the modes for a substantial number of environments worldwide and particle metrics, which suggests that modality in particle size distributions may be a parameter that has potential to be used in the development of PM_1 air quality guidelines and standards. Based on these analyses, implications for choosing different mass standards for airborne particulate matter are discussed in the paper.

1. Introduction

Various aspects are considered when developing ambient air quality standards of which the most important are the exposure-response relationship and the characteristics of the pollutant, which determine the exposure. Size distribution is one of the key characteristics of ambient particulate matter, on one hand related to particle formation and post-formation processes and, on the other hand, determining the fate of particles in the air and

the likelihood of their deposition in the human respiratory tract. Current ambient air quality standards for $PM_{2.5}$ and PM_{10} fractions are based in part on a scientific basis, but also in part on the data and limitations of the size ranges measured by equipment at the time of setting the standards. $PM_{2.5}$ and PM_{10} fractions are mass concentration of particles with aerodynamic diameters smaller than 2.5 and 10 μm, respectively. PM_1 is the mass concentration of particles with aerodynamic diameters smaller than 1 μm.

Size-selective inlets which remove particles that exceed a specific aerodynamic diameter are characterised by sampling effectiveness curves which show the fraction of particles passing through as a function of aerodynamic diameter. Sampling effectiveness is summarised by the 50% cut-point (relating to the diameter that represents half the particles passing through the inlet) and includes a slope function, representing the contribution from different particle sizes above and below the 50% cut-point, because an exact sharp cut-point cannot be achieved in practice (Baron and Willeke 2001).

The $PM_{2.5}$ fraction is sometimes referred to as fine particles, while the difference between PM_{10} and $PM_{2.5}$ is sometimes referred to as coarse particles. Particles larger than 10 μm tend to have atmospheric lifetimes that are relatively short (Harrison et al. 2000) and are of lesser significance from the health point of view since they are mostly removed by the nose. Prior to setting the $PM_{2.5}$ standard, the US EPA conducted an extensive examination of the available data on particle size distributions. The Air Quality Criteria for Particulate Matter (EPA, 1996) contains a comprehensive discussion of the relative merits of PM_1 and $PM_{2.5}$. The decision by the US EPA to introduce 2.5 μm as the upper end of the boundary for fine particles and as a basis for a standard (Reference US Federal Register) was strongly influenced by the fact that the available epidemiological data at the time were obtained using $PM_{2.5}$ measurements (Dockery et al. 1993).

An alternative approach in classification of the particles for the purpose of developing control measures, is to consider location of the modes in particle size distributions, which relate to the contribution from different pollution sources. A mode may be defined as a peak in the lognormal function of the number or mass distribution of an atmospheric aerosol (John, 1993). A number of investigations into the variation of the aerosol size spectrum over a variety of size intervals have been made. Three terms have been introduced for atmospheric aerosol size distribution in terms of modal diameters; these classifications focused on particle size and production mechanism and were the nucleation mode (< 0.1 μm), accumulation mode (0.1-1 μm) and coarse particle mode (> 1 μm) (Jaenicke, 1993).

However it is acknowledged that the location of the modes generally depends on the metric being referred to, such as particle number, surface area, volume or mass, and modes will also change depending on the mathematical transformation method used. For example, Whitby's model of particle volume size distribution (1978) was based primarily on atmospheric aerosol number distributions in the size range 0.01-6 μm, which when transformed to volume distributions, revealed three modal size ranges, with the nuclei mode (< 0.1 μm), the accumulation mode (0.1-2 μm) and coarse particle mode (> 2 μm) (Baron and Willeke 2001). More recently, studies with instruments extending the small size limit to 3 nm have shown that the nuclei mode needs to be separated into a nucleation mode (< 0.01μm) and an Aitken nuclei mode (0.01-0.1μm) (USEPA 2004).

In environments affected by anthropogenic influences most of the nucleation mode particles originate either from the condensation and coagulation of hot, highly supersaturated vapours released during combustion or arise from the condensation and coagulation of low vapour pressure materials formed in the atmosphere by photochemically initiated processes. Coagulation and heterogeneous nucleation tend to

accumulate the aerosol in the accumulation mode. Nucleation, Aitken, and accumulation modes contain soot, acid condensates, sulfates and nitrates, as well as trace metals and other toxins. Most anthropogenic pollution sources are combustion-related and generate particles with diameters < 1 μm (Jamriska and Morawska 2000). Submicrometer particles (diameters < 1 μm) represent most particle matter that is dispersed in urban environments in terms of particle number concentrations (Morawska et al. 1998; Nazaroff et al. 1990). Almost all particles in the coarse particle mode originate from natural and anthropogenic mechanical processes, including grinding, breaking and wear of material and dust resuspension.

The currently accepted division between fine and coarse particles of 2.5 μm does not follow the natural division between modes attributable to different types of sources. Instead, it tends to cut through the mode originating from mechanical processes. It has been shown, however, that there is usually a clear separation between the accumulation and coarse modes around 1 μm or somewhat above, where the mass of particles belonging to these two modes is at a minimum (Lundgren and Burton 1995). Therefore the rationale behind the classification of one micrometer as a division between fine and coarse particles in particle mass and particle volume size distributions would be that it constitutes a natural division between particles generated mainly from combustion and photochemical processes and particles generated from mechanical processes. Obviously this definition, as any, would still be somewhat arbitrary, as nature itself does not provide a perfect division.

Knowledge and understanding of the presence and location of modes in particle distributions is of importance not only for understanding the mechanisms of atmospheric processes, but also, importantly, for exposure and risk assessment, particularly for setting standards and guidelines for air quality. The disparity between what the standards divide

into fine and coarse particles and what nature divides into modes originating from different sources may make control of particles more difficult and in fact may also be less desirable from the health point of view.

The aim of the work reported in this paper was to analyse the available information on modal locations in ambient particle size distributions and, based on this, to explore the potential for PM_1 as an effective mass standard together with PM_{10} in controlling contributions from different types of air pollution sources.

2. Methods and Techniques

The analysis conducted within the scope of this work was divided into two steps. Firstly, characteristics of the modality in particle size distributions for a range of environments in South East Queensland, Australia were investigated to examine the relationship between fractional contribution of mass from different modes in particle size distribution (and thus from different sources) to PM_1, $PM_{2.5}$ and PM_{10}. South East Queensland was chosen because for this environment the authors have detailed information available on particle size distributions, with thousands of spectra collected. The conclusions as to modality of particle size distributions reached by Morawska et al., (1999), as well as the averaged size distributions obtained, served as bases for the analyses presented in this paper.

Particle modal characteristics, their dependence on local conditions in South East Queensland and their variability with time were reviewed by Morawska et al., (1999). This paper also provided a detailed analysis of the modal characteristics of over 6 000 particle size spectra collected over a period of three years for a range of environments, including marine, modified background, suburban background, traffic influenced, urban influenced and vegetation burning. Details concerning the classification of these environments are provided in Morawska et al. (1999). Measurements of size distributions in the size range 0.016 to 30 μm were conducted using SMPS and APS

instrumentation. Spectra corresponding to one sample were combined, normalised and smoothed using a chi-square fitting procedure to give one distribution, and Kolmogorov-Smirnov (K-S) tests were used to compare measured aerosol size distributions (For details see Morawska et al. 1999). The aim of the analysis was to combine the distributions from two instruments measuring submicrometer and supermicrometer particle size distributions for the calculation of the volume size distributions and to allow interpretation of the modal characteristics for each environment studied. The focus of that work was on source identification and identification of the relationship between the sources and size distribution of particles generated. For each environment there was a clear division between the accumulation and coarse modes, but not between the nucleation and accumulation modes. As the densities of the aerosols were not known, only volume and not mass distributions were calculated. As there is, however, a direct correlation between mass and volume distributions, where density acts as a scaling factor, modality displayed by volume and mass distributions are the same.

For each distribution referred to above, in this work, the fractional contribution of N+A (nucleation and accumulation) and C (coarse) modes to volumes of PM_1, $PM_{2.5}$ and PM_{10} were calculated. Ultrafine particles (diameters of < 0.1 μm) tend to dominate particle number and make a significant contribution to surface area but little to mass, with the cube dependence of volume (and therefore mass) resulting in significantly different particle size distributions for particle number and mass distributions (Harrison et al. 2000). In simple terms, it is likely that the majority of particle number is in the transient nucleation and Aitken modes, particle surface area in the accumulation mode, and volume and mass divided between the accumulation and coarse particle modes (Harrison et al. 2000).

The relative contributions were calculated by summing the volumes under the peaks of the modes with the boundary between the accumulation and coarse modes being taken as the sharp visible division on the figures. The total volume of the individual modes was not calculated, which could have been done by extrapolating the curves that describe the mode down to zero on the horizontal axis or fitting a statistical mixture model. Instead the contributions to $PM_{1,}$ $PM_{2.5}$ and PM_{10} were calculated assuming sharp cut-offs. While due to the limitations in the measurement techniques these cut-offs are not sharp, it was considered that for the purpose of the assessment conducted in this work this assumption would not affect the overall outcome of the assessment, but would significantly simplify the calculations. Moreover, where modes overlap, the concentration levels are usually a few orders of magnitude lower than in the peaks and therefore the contribution from the volumes not included was considered to be negligible. Fractional contributions to the modes were calculated by integrating the area under the curve using the trapezoidal rule. The trapezoidal rule takes account of the different width of the x-axis in each bin, and may be applied to either the original data scale or the log scale. Log scale values were used in our calculations and calculations were undertaken using Origin (Version 6.0).

Secondly, an analysis of modal locations reported in international literature was conducted to determine whether a clear and distinct separation occurs in the log-transformed data between the modes around 1 μm, in different environments and for different metrics. This was evaluated by constructing a 95% confidence interval for the mean of those modal values lying below 1 μm, and a second 95% confidence interval for the mean of those values lying above 1 μm. The means of the two groups were asserted to be significantly different if these two confidence intervals did not overlap. Moreover, the value of 1 μm was determined to be an effective threshold if it separated the two intervals, so that it was larger than the confidence interval for the smaller mean, and smaller than the confidence interval for the larger mean. This was evaluated by testing

for the existence of two modal groups with significantly different means, as determined by non-overlapping 95% confidence intervals. This analysis was used to ascertain which environments and metrics may possibly be suited to PM_1 standards.

3. Results and discussion

Contribution of the modes in South East Queensland to PM_1, $PM_{2.5}$, PM_{10}

Figure 1 presents averaged size distributions for different types of environments in South East Queensland in terms of both number and volume size distribution. To enable the distinctions between the modes and identification of the size boundaries of the modes, both types of spectra are presented in double logarithmic scale. A vertical line shows the location of the division according to the boundary of $PM_{2.5}$ and coarse particles.

A general conclusion that can be made from inspection of the distributions presented in Figure 1 is that in all of the environments there is a good separation between accumulation and coarse particle modes, but that this separation occurs at or below 1 μm. Harrison et al. (2000) found a similar separation at around 1 μm in measured particle size distributions from suburban Birmingham, United Kingdom in terms of number, surface area and volume. It can be seen in the South East Queensland environments that in all cases the division at 2.5 μm cuts across the coarse particle mode, close to its peak.

Inspection of the spectra presented in Figure 1 reveals that for traffic influenced aerosols as well as for urban influenced, suburban background and modified background aerosols both number and volume distributions are bimodal, with the majority of particle number being associated with the fine particle mode (nucleation, Aitken and accumulation regions, N+At +A), while most of the mass is associated with the coarse particle mode (C). Since our instruments only extended to 16 nm, we do not have information on the nucleation mode and the Aitken mode and the accumulation mode are not clearly separated in all size distributions. Similarity between the modal locations in the N+At and

A region in these environments leads to the conclusion that in this urban environment automobile exhausts are the major contributors.

The coarse particles, on the other hand, may more likely result from a number of different sources and not just the predominant road dust source for aerosols sampled adjacent to the freeway, as indicated by differences between the shapes of the size distribution curves. For example, the distributions for modified background aerosols are considered representative of the influences by biogenic sources, with the broad width of the coarse particle mode being a reflection of the presence of particles originating from plant emissions in the aerosols. The existence of several more peaks in the suburban aerosol is likely the result of several background aerosol sources, which in urban type locations are usually masked by the presence of much stronger sources, such as traffic emissions.

There is also close similarity between the shapes of the size distribution curves of vegetation burning influenced aerosols and the traffic and urban aerosols in South East Queensland. There is a difference, however, in the width of the modes, with the N+A mode of the vegetation burning influenced aerosol being at a larger particle size than the other two aerosols typically encountered in urban environments. There are a number of peaks present within the N+A modes of the marine influenced aerosol as presented in Figure 1. They include free troposphere nuclei mode, effects related to the influence of cloud processing of coagulating nuclei and the sea salt component of marine aerosols. While the majority of the particles in the number size distribution are smaller than 1 μm diameter, the majority of the volume is in fact occupied by particles with diameters greater than 1 μm.

For each of the distributions presented in Figure 1, fractional contribution of N+A and C modes to the volumes of PM_1, $PM_{2.5}$ and PM_{10} was calculated, assuming, as discussed

above, sharp cut-offs of 1, 2.5 and 10 μm. These contributions are shown in Table 1 and form the basis of our study conclusions. The most obvious conclusion from Table 1 is that PM_{10} volume in all environments, except vegetation burning, can be attributed mainly to particles from the coarse mode (C), that is, particles generated from mechanical processes. Contribution from combustion processes to PM_{10} is negligible. Volume from N+A modes for vegetation burning contributes about 50% to PM_{10} volume. Similarly to PM_{10}, in most of the environments the Coarse (C) mode has the strongest contribution to $PM_{2.5}$ volume. However, in traffic influenced and vegetation burning the contribution from N+A is substantial. In the case of vegetation burning, N+A volume has a stronger contribution to $PM_{2.5}$ volume compared to C volume and to its contribution to PM_{10} volume. Contribution from N+A mode volume to PM_1 is dominant for traffic influenced, vegetation burning, marine influenced and modified background.

Modal locations in the published literature

The review of published studies revealed 605 modes reported for particle number, surface area, volume and mass size distributions. Since access to the data used by other authors was not available, the examination focused on the location of these reported modes. Moreover, for the purposes of this study, only modal location values ≤ 10 μm were extracted. Of the 605 modes identified, five occurred at ≥10 μm in particle volume and were not included in the review, leaving a total of 600 examined in our study. Particle concentrations and their relative variations were not considered in this study. The published values spanned diverse environments, and included background, central european aerosol, desert, fires, forest, high alpine, marine and modified marine, modified background, north-west Himalayas, rural/continental, suburban, traffic-influenced, urban-influenced, urban background and vegetation burning environments. Tables 2-5 present listings of the international studies reviewed.

Figure 2 presents a compilation of all 600 modal location values from the analysis for a range of environments and metrics. Modal location value ranges for the different metrics spanned from 0.006 to 3 μm for number; 0.02 to 3.5 μm for surface area; 0.008 to 10 μm for volume and 0.06 to 7.8 μm in mass particle size distributions. Approximately 98% of number modal location values occurred at ≤ 1 μm. Surface area modal locations showed a similar pattern to mass but were shifted to the right, to the larger size ranges.

Three conclusions can be made from inspection of the results presented in Figure 2. Firstly, it can be seen that there is a clear and distinct separation between the modes at 1 μm for all worldwide environmental data reviewed for surface area, volume and mass size distributions. The one exception is a volume size distribution mode identified in marine and modified marine in Tasmania at 1 μm by Gras and Ayers (1983) where the salt component was found to comprise more than 95% of the total volume. Secondly, it can be seen in Figure 2 that clusters of modal values appear for each metric. Finally, the figure shows that number and volume size distribution modal location values for South East Queensland generally fell within the modal size ranges reported for the worldwide environments.

The effect of relative humidity on particle size under certain circumstances is important and has been the topic of many investigations. For example, Mobility Analysers can change relative humidity conditions during sampling, and in many cases heat the sampled air sufficiently to reduce the size of the particles. Mass size distributions measured at high relative humidity or in clouds or fog show considerable fine particulate matter above 1μm. However, the papers reviewed by this study comprised a very wide range of conditions, including studies related to high humidity conditions. Overall this has not had an impact on the inferences. In fact the study region in South-East Queensland experiences an annual average relative humidity of between 60-73% in the mornings and

49-60% in the afternoons. Therefore it appears that in the majority of cases (or under most circumstances) humidity is not a factor changing the location of the mode according to the conclusions discussed here. This is an important conclusion in relation to considerations in setting standards, as these need to account for the majority of cases, especially in relation to anthropogenic contributions.

Separation between modal location values in mass and volume particle size distributions at around 1 μm

Of the 600 modal values examined in this study in particle number, surface area, volume and mass size distributions 87 modes (15%) were clustered closely on either side of 1 μm, and five modes were found at exactly 1 μm. As clearly indicated in Figure 2, the modal values formed two distinct subgroups above and below 1 μm. The upper 95% confidence bound of the smaller mean, and the lower 95% confidence bound of the larger mean for particle volume and mass size distributions, are displayed by vertical dashed lines in Figure 2. The lack of overlap between these confidence intervals is apparent, confirming the existence of two groups with statistically different means. The confidence intervals were calculated for modal value clusters at between 0.197 and 0.5 μm and 1.84 and 8 μm in volume; and 0.43 and 0.65 μm and 3.16 and 5.06 μm in mass size distributions. To facilitate comparison between South East Queensland and modal values reported elsewhere in the world, confidence intervals for South East Queensland modal values in volume size distribution were not calculated.

When considering all the modal location values depicted in Figure 2, a distinct gap was found between the location of the modes at both below and above 1 μm. This distinct gap occurred at between 0.65 and 2 μm in mass particle size distributions; between 0.3 and 2.2 μm in surface area; between 0.5 and 1.8 μm in volume and between 0.8 and 1.2

μm for number. It should be noted that two distinct modal location values present at 1 μm and 2 μm in Figure 2 related to marine environments. These were a mode found at 2 μm in particle mass, which related primarily to sea salt particles in the remote marine boundary layer in the high Arctic over the central Arctic Ocean (Hillamo et al. 2001) and a mode at 1 μm in volume in an undisturbed marine environment in the southern mid-latitudes, west coast of Tasmania, Australia, where the salt component made up more than 95% of the total volume (Gras and Ayers 1983).

4. Conclusions

The relation between fractional contribution to volume and mass from different modes in the particle size distribution (and thus from different sources) to PM_1, $PM_{2.5}$ and PM_{10} was examined in this paper, based on a large body of data on ambient particle size distributions from the measurements conducted in South East Queensland, Australia. The conclusions from the analyses in relation to developing air quality regulations are as follows. Firstly, PM_{10} measurements provide information almost entirely on particles generated from mechanical processes and belonging to the coarse mode. In an urban environment this could also mean particles resuspended by the vehicular traffic and mechanical wear and tear of the tyres, but not emitted from motor vehicles.

Secondly, $PM_{2.5}$ measurements (coarse mode) also provide information mainly on particles generated by mechanical processes, but the contribution from combustion process modes (nucleation and accumulation modes) becomes significant for some environments. Thus interpretation of $PM_{2.5}$ data could become very complex in order to distinguish the contribution from different types of sources. It follows that the application of this $PM_{2.5}$ parameter, as a basis for standards may not adequately facilitate control of particle emissions and concentrations.

Thirdly, PM_1 measurements (nucleation and accumulation modes) provide very good information about contributions from combustion processes and enable a much better distinction to be made between combustion and mechanically generated aerosols. It would thus appear that PM_1 and PM_{10} mass standards would be most desirable from the legislation point of view.

The review of 600 modal location values for particle number, surface area, volume and mass size distributions in a wide range of environments worldwide revealed a clear and distinct separation around 1 μm. A similar separation was found in all the South-East Queensland environments examined in terms of the separation between accumulation and coarse modes for volume and number size distributions, which occurred at around 1 μm.

We conclude that examination of the location of the modes in particle size distributions has potential as a basis for developing air quality standards and guidelines as modes provide useful information about contributions from different pollution sources and particle mechanisms. Therefore, based on both the local South East Queensland study and the other studies conducted around the world, it is concluded that PM_1 and PM_{10} offer greater potential as a combination for particle mass standards than the current mass standards of $PM_{2.5}$ and PM_{10}.

Two additional points need to be discussed when considering a PM_1 standard. Firstly, while at the moment very little data are available on PM_1 concentrations, there are measurement technologies available to undertake these measurements, which are very similar to those used for $PM_{2.5}$ monitoring. Secondly, in addition to particle mass concentration standards, future legislations may also consider number concentration standards, which would be focused on submicrometer or even smaller, ultrafine particles. In urban areas, for example, motor vehicles are the major emitter of ultrafine particles,

which are very small and prolific in terms of particle number, but have negligible mass. The rapid progress in the monitoring technologies available to measure particle number concentration currently makes such measurements possible. While this paper considered only the rationale for the most advantageous combination of particle mass standards from the legislative point of view, more discussion should be conducted to consider the best combination of particle mass and number concentration standards.

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Tables and Figures

Table 1 Percent contribution of N+A and C modes by mass to PM_1, $PM_{2.5}$ and PM_{10} in South East Queensland, Australia

Environment type	PM1 % contribution (by mass)		PM2.5 % contribution (by mass)		PM10 % contribution (by mass)	
	N+A	C	N+A	C	N+A	C
Traffic Influenced	99	1	61	39	24	76
Urban Influenced	49	51	3	97	< 1	> 99
Vegetation burning	100	0	90	10	52	48
Marine influenced	82	18	2	98	<1	>99
Modified background	88	12	13	87	< 1	> 99
Suburban background	38	62	1	99	< 1	> 99

Table 2 International literature reviewed to identify the location of the modes in a number of different environments worldwide for particle number size distributions

Condition	Researchers	Particle size range measured, μm)	Location
Central European Aerosol	Neususs et al. 2002	0.003-10	Leipzig and Berlin, Germany
Continental background	Birmili et al. 1999 & 2001	0.003-0.8	Melpitz, Germany
Continental background	Wiedensohler et al. 2002	0.003-0.8	Melpitz, Germany
Forest	Makela et al. 2000	0.003-0.5	Southern Finland
Forest	Tunved et al. 2005	0.01-0.5	Hyytiala, Matorova Station, Varrio, Finland
High Alpine	Weingartner et al. 1999	0.018-0.75	Jungfrauhoch, Switzerland, 3580m
Marine	Heintzenberg et al. 2004	0.0031-0.65	Cape Grim, Australia
Marine	Heintzenberg et al. 2004	0.0031-0.79	Sagres, Portugal
Marine	Heintzenberg et al. 2004	0.003-0.9	N/S Atlantic, Indian Ocean, Pacific, Yellow Sea, Sea of Japan
Marine & modified marine [a]	Morawska et al. 1999	0.016-30	Brisbane, Australia
Marine & polluted air masses	O'Dowd et al. 2001	0.005-150	Mace Head, Ireland
Modified background	Morawska et al. 1999	0.016-30	Brisbane, Australia
Rural	Tunved et al. 2005	0.01-0.452	Aspvreten, Sweden
Suburban	Hussein et al. 2005	0.003-0.6	Finland
Suburban background	Morawska et al. 1999	0.016-30	Brisbane, Australia
Traffic-influenced	Morawska et al. 1999	0.016-30	Brisbane, Australia
Traffic-influenced	Pirjola et al. 2004	0.007-10	Helsinki, Finland
Traffic-influenced	Rosenbohm et al. 2005	0.0107-10 northside), 0.0202-10 (southside)	Heidelberg, Germany
Traffic-influenced	Zhu et al. 2002 a,b & 2004	6-220	Los Angeles, USA
Traffic-influenced	Zhu et al. 2006 [a]	7-300	Los Angeles, USA
Transition zone between continental boundary layer and free troposphere	Van Dingenen et al. 2005 [b]	0.006-10	Monte Cimone Observatory, Italy
Urban	Hussein et al. 2004	0.008-0.4	Kumpula and Siltavuori, Finland
Urban	Hussein et al. 2005	0.003-0.6	Siltavuori and Pasila, Finland
Urban	Monkkonen et al. 2005	0.003-0.8	New Delhi, India
Urban	Wehner et al. 2002	0.003-0.8	Leipzig, Germany
Urban	Wiedensohler et al. 2002	0.003-0.8	Leipzig, Germany
Urban	Fine and Sioutas 2004	0.0141-2.5	LA Basin, USA
Urban	Salma et al. 2002	0.01-10	Budapest, Hungary
Urban	Morawska et al. 1999	0.016-30	Brisbane, Australia
Vegetation burning	Morawska et al. 1999	0.016-30	Brisbane, Australia

[a] Night-time data

Table 3 International literature reviewed to identify the location of the modes in a number of different environments worldwide for particle surface area distributions

Condition	Researchers	Particle size range measured, μm)	Location
Transition zone between continental boundary layer and free troposphere	Van Dingenen et al. 2005 [a]	0.006-10	Monte Cimone Observatory, Italy
Urban	Salma et al. 2002	0.01-10	Budapest, Hungary
Vegetation burning	Jayaratne & Verma 2001	0.1-5	Gaborone, Botswana, Southern Africa

. [a] Night-time data only

Table 4 International literature reviewed to identify the location of the modes in a number of different environments worldwide for particle volume size distributions

Condition	Researchers	Particle size range measured, μm)	Location
Background	Hidy 1975	0.015-30	Southern California, USA
Central European Aerosol	Neususs et al. 2002	0.003-10	Leipzig and Berlin, Germany
Desert	Hidy 1975	0.015-30	Southern California, USA
Marine	Hidy 1975	0.015-30	Southern California, USA
Marine and modified marine [a]	Hoppel et al. 1990	0.006-2.2	Wallops Island, USA
Marine and modified marine [a]	Gras and Ayers 1983	0.0025-5	Tasmania, Australia
Marine and modified marine [a]	Porter and Clarke 1997	0.17-7.5	Tasmania, Australia
Marine and modified marine [a]	Porter and Clarke 1997	0.17-7.5	Hawaii, USA
Modified marine [a]	Morawska et al. 1999	0.016-30	Brisbane, Australia
Marine and polluted air masses	O'Dowd et al. 2001	0.005-150	Mace Head, Ireland
Modified background	Morawska et al. 1999	0.016-30	Brisbane, Australia
Suburban	Meszaros 1977	0.020-100	Budapest, Hungary
Suburban background	Morawska et al. 1999	0.016-30	Brisbane, Australia
Traffic-influenced	Hidy 1975	0.015-30	Southern California, USA
Traffic-influenced	Morawska et al. 1998	0.016-30	Brisbane, Australia
Transition zone between continental boundary layer & free troposphere	Van Dingenen et al. 2005 [b]	0.006-10	Monte Cimone Observatory, Italy
Urban	Morawska et al. 1999	0.016-30	Brisbane, Australia
Vegetation burning	Jayaratne & Verma 2001	0.1-5	Gaborone, Botswana Southern Africa
Vegetation burning	Morawska et al. 1999	0.016-30	Brisbane, Australia

[a] Modified marine in these cases refers to marine aerosol influenced by continental air parcels. [b] Night-time data only

Table 5 International literature reviewed to identify the location of the modes in a number of different environments worldwide for particle mass size distributions

Condition	Researchers	Particle size range measured, μm)	Location
Himalayas	Gajananda et al. 2005	0.08-9	North-west Himalayas, India
Marine	Hillamo et al. 2001	0.045-10	High Arctic, remote boundary layer
Rural	Berner et al. 2004	0.06-16	Vienna, Austria
Traffic	Berner et al. 2004	0.06-16	Vienna, Austria
Urban	Berner et al. 2004	0.06-16	Vienna, Austria
Urban	Salma et al. 2002	0.01-10	Budapest, Hungary
Urban	Salma et al. 2005	0.05-10	Budapest, Hungary

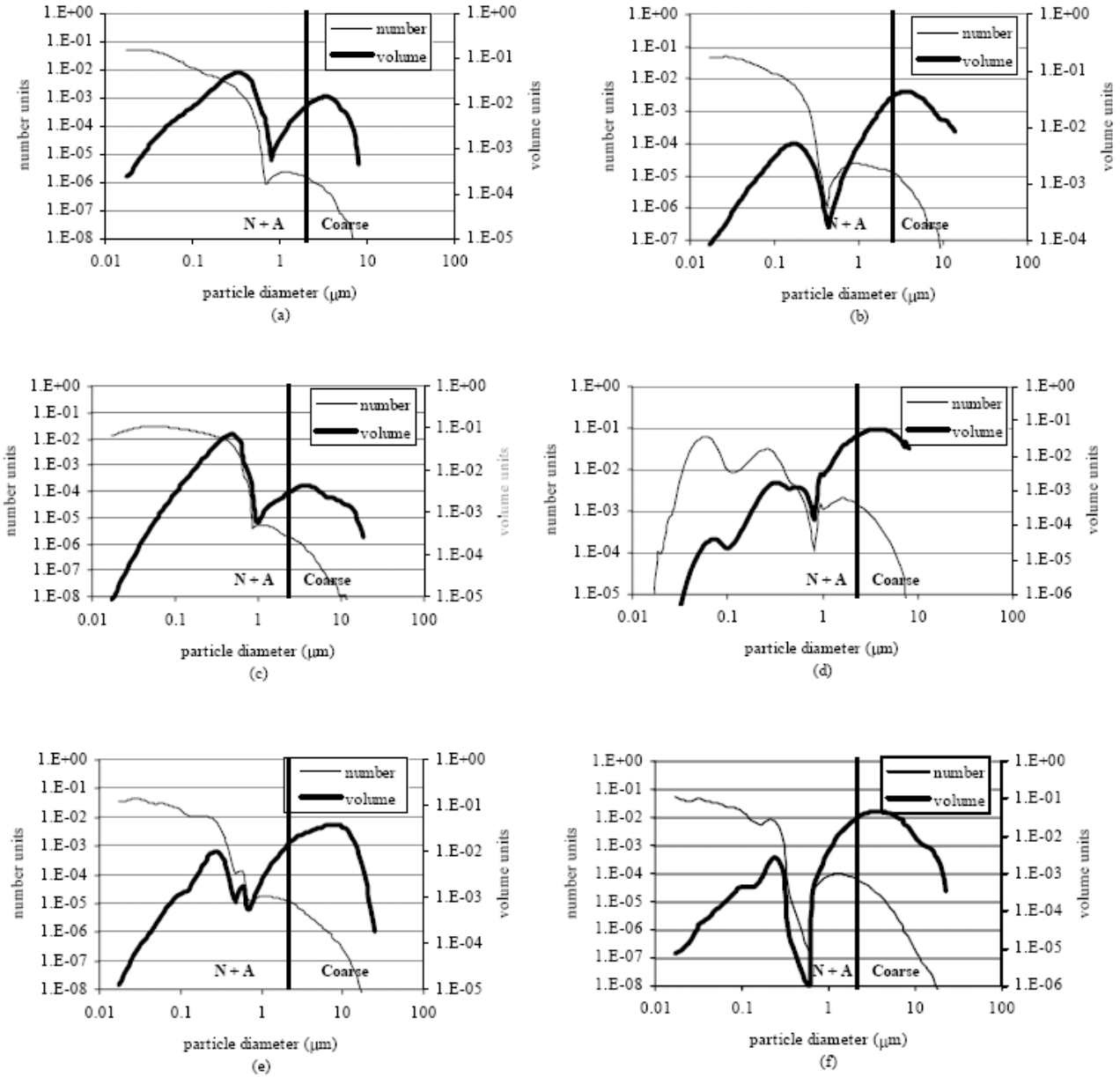

Figure 1. Normalised number and volume size distributions in South East Queensland, Australia traffic influenced aerosol (b) urban influenced aerosol (c) vegetation burning influenced aerosol (d) marine influenced aerosol (e) modified background aerosol (f) suburban background aerosol. N + A (nucleation and accumulation modes), Coarse (coarse mode).

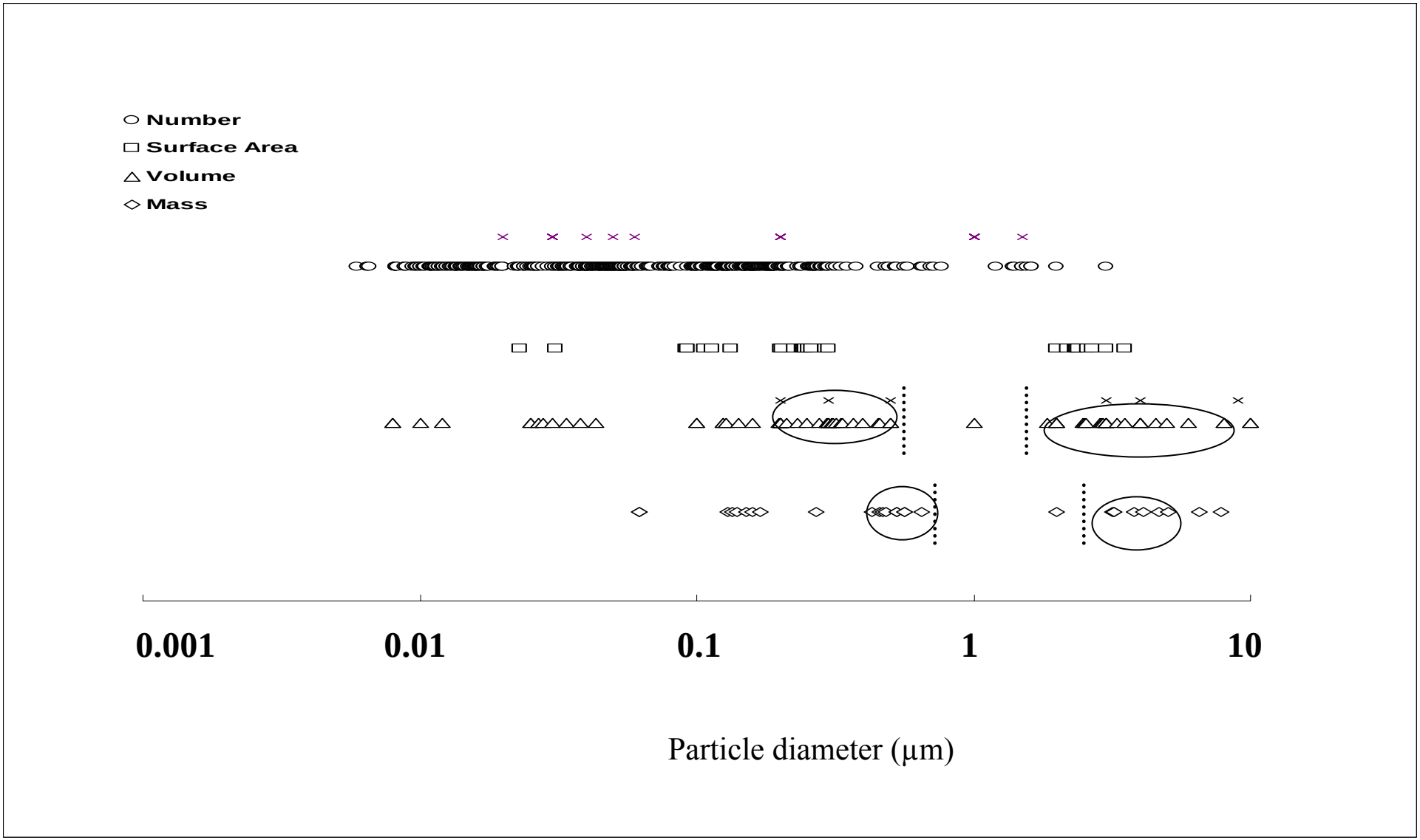

Fig. 2 Published modal location values relating to particle size distributions for South East Queensland, Australia (marked x) and for a range of environments worldwide and metrics (n=600). Vertical dashed lines indicate the 95% Confidence interval upper bounds for modal value clusters to the left of 1 μm and 95% Confidence interval lower bounds for modal value clusters to the right of 1μm in particle volume and mass size distributions, these modal value clusters are circled above.